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Comment on "Brillouin Study of the Quantization of Acoustic Modes in Nanospheres"

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In a recent Letter¹, M. H. Kuok et al report interesting inelastic light scattering measurements of acoustic vibrations in three-dimensional ordered arrays of unembedded SiO₂ nanospheres a few hundreds nanometer in diameter. The use of a Brillouin spectrometer makes it possible to record spectra with several scattering lines within 1 cm⁻¹ of the Rayleigh line when good Raman spectrometers can't go below a few cm⁻¹. Their samples also had a tight size and shape particles distribution resulting in narrow scattering lines.

However their analysis errs in calculating the energy of the $l = 0$ spheroidal mode. This same error is recurrent in the literature^{2,3,4}. Some of them have since corrected this⁵. The original Lamb paper⁶ was correct and an explanation is available elsewhere⁷.

This error results in additional energies which don't correspond to any vibration eigenmode. It gives a wrong energy for the lowest $l = 0$ spheroidal mode comparable to the one of the lowest $l = 2$ spheroidal mode. Therefore the discussion is based on wrong line assignment.

In this letter, the sound velocities of silica reported in the literature were not used. Instead, a fit of the observed frequencies was made to determine them. This is not common in low-frequency Raman related papers. Because of the previous error, the fit was made on wrong assumptions.

In table I, we provide corrected frequencies with published sound velocities⁸ and with Kuok et al's fitted velocities. These values correspond to the letter's coefficients in equation 3. We believe the mode assignment is not as simple as the one reported in the letter.

	l	n			
		1	2	3	4
published	0	4.232	10.925	16.819	22.651
sound velocities	2	3.146	5.376	9.209	10.982
fitted	0	4.012	10.083	15.520	20.905
sound velocities	2	2.798	4.909	8.446	9.924
Kuok et al.	0	(2.617)	4.017	(6.343)	9.804
	2	2.796	4.098	8.427	

TABLE I: Calculated mode frequencies (GHz) for a 1 μ m diameter SiO₂ free sphere. Values in parenthesis result from the calculation error for $l = 0$ modes.

Moreover, a few points in this letter are misleading. From the introduction, it seems that this work is the first report of light scattering from free (non-embedded) nanoparticles. However, a few papers have already reported such measurements in TiO₂^{9,10} and SnO₂^{11,12} powders.

The authors also completely disregard any effect associated to the periodicity of the particle's network and the particle's interconnections. Recent work by Lazarenkova et al.¹³ shows that "the acoustic-phonon dispersion in the quantum dot superlattice undergoes strong modification, which leads to emergence of quasi-optical branches". It is not obvious that the zero stress boundary condition is more suitable in this system than for matrix-embedded particles. This might be the reason why the measured energies are not in good agreement with the free sphere model.

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